

Thermal Conductivity in the Triangular-Lattice Antiferromagnet $\text{Ba}_3\text{CoSb}_2\text{O}_9$

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Abstract. We have measured the thermal conductivity along the ab -plane and c -axis, κ_{ab} and κ_c , of the $S = 1/2$ triangular-lattice antiferromagnet $\text{Ba}_3\text{CoSb}_2\text{O}_9$ single crystals in magnetic fields parallel to the ab -plane up to 14 T, in order to investigate the change of the magnetic state. It has been found that κ_{ab} and κ_c are enhanced suddenly at low temperatures below 3.8 K, where an antiferromagnetic order of the 120° structure in the ab -plane occurs in zero field. On the other hand, the magnetic-field dependence of κ_{ab} and κ_c at 2.8 K show increases around 8 T, where $\text{Ba}_3\text{CoSb}_2\text{O}_9$ undergoes the $1/3$ magnetization plateau in magnetic fields parallel to the ab -plane. Furthermore, the magnetic-field dependence of κ_{ab} and κ_c show dips around 13 T which is the middle of the $1/3$ magnetization plateau. These dips may be due to a possible change of the magnetic state.

1. Introduction

Recently, thermal conductivity in quantum spin systems has attracted considerable interest because it is closely related to the spin state. First, large thermal conductivity due to magnetic excitations (κ_{spin}) has been observed in one-dimensional quantum spin system Sr_2CuO_3 [1, 2, 3] and SrCuO_3 [3, 4]. Moreover, since magnetic excitations scatter phonons, a change of magnetic state affects the thermal conductivity due to phonons (κ_{phonon}). This indicates that thermal conductivity measurements is useful probe to investigate the change of magnetic state and phase transitions. For example, it is reported that spin gap is related to κ_{phonon} in $\text{SrCu}_2(\text{BO}_3)_2$ [5, 6]. Since the number of magnetic excitations which scatter phonons is determined by spin gap, the spin-gap formation and the reduction of spin gap lead to increase and decrease of κ_{phonon} , respectively.

In $\text{Ba}_3\text{CoSb}_2\text{O}_9$, Co^{2+} spins with the spin quantum number $S = 1/2$ form an uniform triangular-lattice in the ab -plane. From the analysis of ESR and magnetization processes results, intralayer antiferromagnetic interaction J and interlayer antiferromagnetic interaction J' are determined to be $J = 18.5$ K and $J' = 0.48$ K [7]. An antiferromagnetic order of the 120° structure in the ab -plane occurs at $T_N \approx 3.8$ K due to weak interlayer interaction J' in zero field [8]. When the magnetic field is applied along the ab -plane, the up-up-down (UUD) structure appears in the magnetic fields above 8 T along the ab -plane. This state causes a $1/3$ magnetization plateau. On the other hand, no magnetization plateau is observed and a cusp appears for the magnetic field along c -axis. These results indicate the easy-plane anisotropy, which is consistent with the collective ESR modes [7]. Magnetic structure of $\text{Ba}_3\text{CoSb}_2\text{O}_9$ in the magnetic fields has been also investigated [7, 9].

It is predicted theoretically that quantum fluctuation stabilize the UUD structure in triangular-lattice antiferromagnets [10]. Experimentally, a $1/3$ magnetization plateau in the $S = 1/2$ triangular-lattice antiferromagnet (TLAF) Cs_2CuBr_4 has been investigated [11, 12, 13]. Since

an magnetization plateau is caused by an energy gap which separates low-energy excitations from the ground state, the UUD state in Cs_2CuBr_4 is gapped state [14, 15]. Thermal conductivity is a useful probe to investigate the change of the magnetic state and the spin gap which stabilizes a $1/3$ magnetization plateau. Moreover, κ_{spin} in trigonal-lattice antiferromagnets is already investigated in $\kappa\text{-(BEDT-TTF)}_2\text{Cu}_2(\text{CN})_3$ [16]. In order to investigate the existence of κ_{spin} and the change of the magnetic state in the UUD state with the $1/3$ magnetization plateau we have measured the thermal conductivity of $\text{Ba}_3\text{CoSb}_2\text{O}_9$ in magnetic fields.

2. Experimental

Single crystals of $\text{Ba}_3\text{CoSb}_2\text{O}_9$ were grown by floating-zone method. The quality of the single crystals was checked by the x-ray back-Laue photography to be good. Thermal conductivity measurements were carried out by the conventional steady-state method. Magnetic fields up to 14 T were applied parallel to the ab -plane.

3. Results and discussion

Figure 1 shows the temperature dependence of the thermal conductivity along the ab -plane and c -axis, κ_{ab} and κ_c , for single crystals of $\text{Ba}_3\text{CoSb}_2\text{O}_9$. κ_{ab} and κ_c show a broad peak around 40 K. While κ_{ab} and κ_c decrease with decreasing temperature below 40 K, they increase below T_N as shown in the inset of Fig. 1. Although temperature dependence of κ_{phonon} usually shows a peak around 10 K, it was not observed around 10 K but around 40 K in $\text{Ba}_3\text{CoSb}_2\text{O}_9$. Magnetic fluctuation arising from frustration may suppress the mean free path of phonons (l_{phonon}). Since the absolute values of the peaks between κ_{ab} and κ_c is much same and the behaviour of the temperature dependences of κ_{ab} and κ_c is isotropic, phonon contribution is dominant in the thermal conductivity. Enhancement of κ_{ab} and κ_c at the temperature below T_N indicates that l_{phonon} is extended due to the suppression of phonon scattering in the ordered state.

Figure 2 shows the field dependences of κ_{ab} and κ_c , $\kappa_{ab}(H)$ and $\kappa_c(H)$, at 2.8 K, 3.4 K,

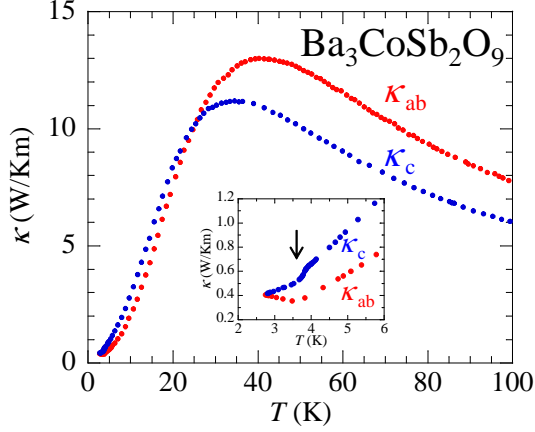


Figure 1. Temperature dependence of the thermal conductivity along the *ab*-plane and *c*-axis, κ_{ab} and κ_c , for single crystals of $\text{Ba}_3\text{CoSb}_2\text{O}_9$. The inset shows an expansion of the graph around $T_N = 3.8\text{K}$.

4 K and 5 K. There is no difference between $\kappa_{ab}(H)$ and $\kappa_c(H)$, which is consistent with the dominant phonon contribution to the thermal conductivity in $\text{Ba}_3\text{CoSb}_2\text{O}_9$. Since phonons can be scattered by magnetic excitations, the behaviour of $\kappa_{ab}(H)$ and $\kappa_c(H)$ attributes the change of the magnetic state. κ_{ab} and κ_c decreased with increasing magnetic field in the 120° structure phase at 2.8 K and 3.4 K, which are below T_N . This decrease is due to the suppression of l_{phonon} , because the number of magnetic excitations increases with increasing magnetic field. κ_{ab} and κ_c increased suddenly around 8 T where $\text{Ba}_3\text{CoSb}_2\text{O}_9$ undergoes the transition from the 120° structure phase to the UUD phase. This increase may indicate that the number of magnetic excitations decreased due to the appearance of the spin gap in the UUD phase and that suppressed l_{phonon} increased. $\kappa_{ab}(H)$ and $\kappa_c(H)$ at 4 K and 5 K show no change in low magnetic field as shown in Fig. 2(b,d), because these temperatures are in the paramagnetic (PM) phase. Then κ_{ab} and κ_c decreased due to the transition from the PM phase to the UUD phase at 5 T and 7 T for 4 K and 5 K, respectively.

While κ_{ab} and κ_c increased at the transition from the 120° structure phase to the UUD phase (Fig. 2(a,c)), κ_{ab} and κ_c decreased at the transition from the PM phase to the UUD phase (Fig. 2(b,d)). This difference may be explained by the number of magnetic excitations. In the 120° structure phase, the number of magnetic excitations is increased due to the magnetic field. At the transition from the 120° structure phase to the UUD phase, the number of magnetic excitations

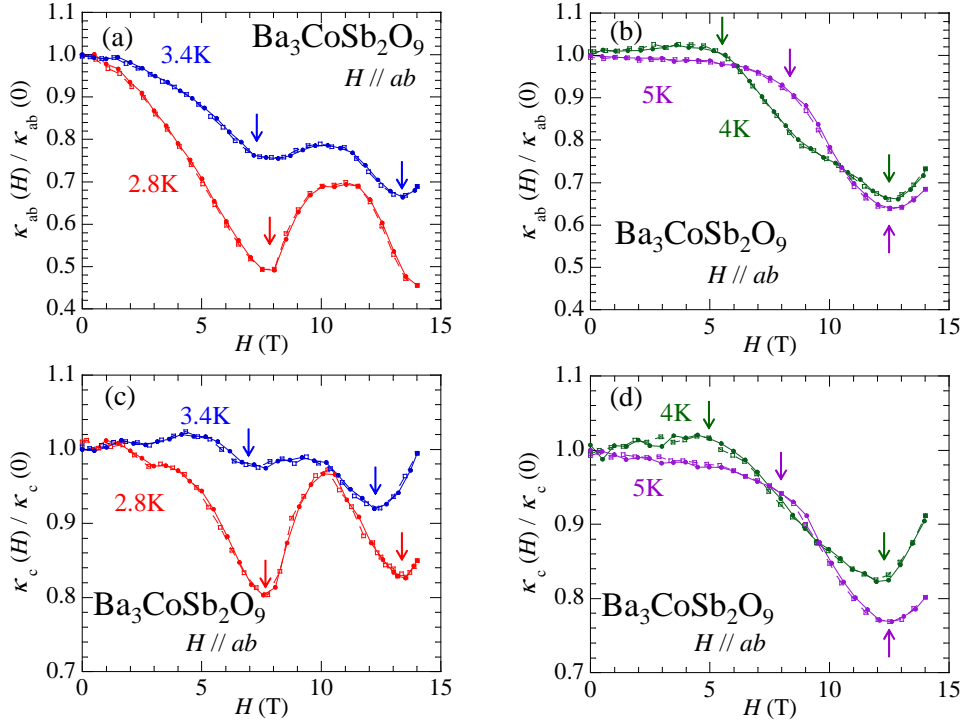


Figure 2. Field dependence of κ_{ab} and κ_c , $\kappa_{ab}(H)$ and $\kappa_c(H)$, (normalized to the 0 T value) for single crystals of $\text{Ba}_3\text{CoSb}_2\text{O}_9$ at 2.8 K, 3.4 K, 4 K and 5 K.

decreases due to the appearance of the spin gap, which leads to the extension of l_{phonon} . On the other hand, in the PM phase, the number of magnetic excitations do not increase with increasing magnetic field. At the transition from the PM structure phase to the UUD phase, the number of magnetic excitations may increase and l_{phonon} may be suppressed. Since the thermal fluctuation is large near the phase boundary in the UUD phase, the extension of l_{phonon} is not observed even in the existence of the spin gap. Arrows in Fig. 2 indicate transition to the UUD phase and the transition fields are shown as \times marks in Fig. 3. Transition fields from the 120° structure phase to the UUD phase are much same with the magnetic phase diagram of ref. [9, 17] shown in Fig. 3. On the other hand, the transition fields from the PM phase to the UUD phase are different. This difference suggests that the magnetic fluctuation appears at higher temperature than the phase boundary by ~ 1 K in the PM phase. Existence of the magnetic fluctuation coincides with

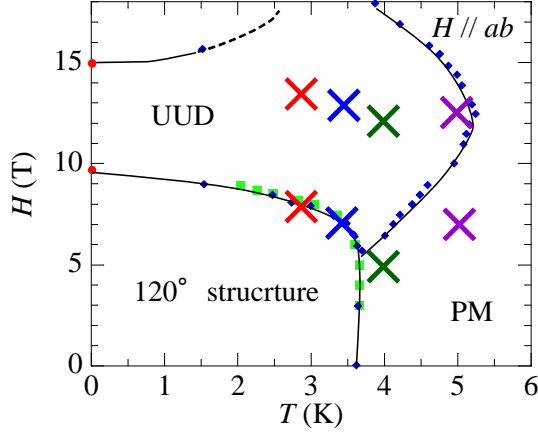


Figure 3. H - T magnetic phase diagram of $\text{Ba}_3\text{CoSb}_2\text{O}_9$ in magnetic fields along the ab -plane. Data were taken from ref. [9, 17]. \times marks are placed at the change in the field dependence of the thermal conductivity shown as the arrows in Fig. 2.

the broad peak in the temperature dependence of the specific heat at the transition from the PM phase to the UUD phase [17].

In high magnetic field above 10 T, $\kappa_{ab}(H)$ and $\kappa_c(H)$ show peaks around 11 T at 2.8 K and 3.4 K (Fig. 2(a,c)). Although these peaks may indicate that the magnitude of the spin gap starts decreasing with increasing magnetic field around 11 T, the origin of these peaks is unclear. Furthermore, $\kappa_{ab}(H)$ and $\kappa_c(H)$ show dips around 13 T at all temperatures 2.8 K, 3.4 K, 4 K and 5 K shown as arrows in Fig. 2. Arrows in Fig. 2 indicate the dips around 13 T and the fields where the dips appear are shown as \times marks in Fig. 3. Figure 3 shows that these dips are placed in the center of the UUD phase. However, no transition is observed in other measurements. Although the field dependences of the specific heat do not show any anomalies which indicate phase transition around 13 T [17], some change may occur. It is necessary to investigate this magnetic state using NMR and neutron scattering measurements.

4. Summary

In order to investigate the existence of κ_{spin} and the change of the magnetic state, we have measured the thermal conductivity along the ab -plane and c -axis, κ_{ab} and κ_c , the $S = 1/2$ triangular-lattice antiferromagnet $\text{Ba}_3\text{CoSb}_2\text{O}_9$ single crystals in magnetic fields. It has been

found that the phonon contribution to the thermal conductivity is dominant due to the isotropic behaviour of the temperature dependence between κ_{ab} and κ_c . Moreover, κ_{ab} and κ_c increased at the temperature below T_N , because the decrease of the number of magnetic excitations leads to the extension of l_{phonon} . In magnetic field along ab -plane at 2.8 K, κ_{ab} and κ_c increased around 8 T, where $\text{Ba}_3\text{CoSb}_2\text{O}_9$ undergoes the UUD phase. This enhancement may be due to the appearance of the spin gap. Furthermore, the magnetic-field dependence of κ_{ab} and κ_c show dips around 13 T which is roughly the midpoint of the 1/3 magnetization plateau. These dips may be due to a possible change of the magnetic state.

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